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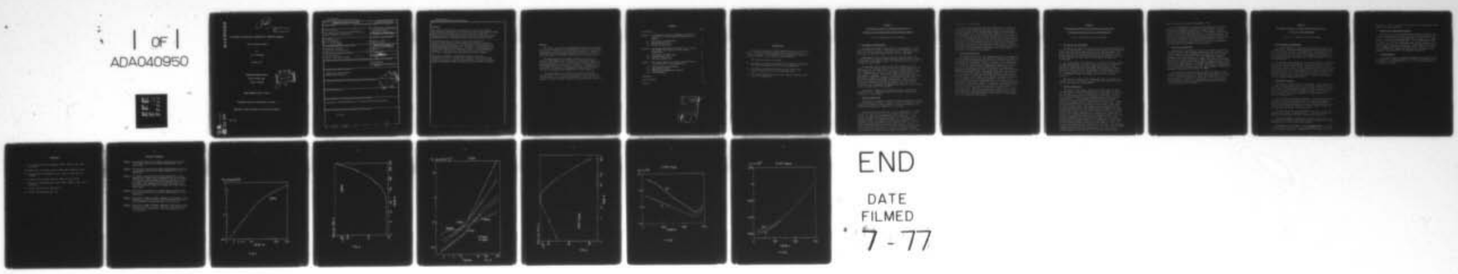

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THE THERMAL AND ELECTRICAL PROPERTIES OF COMPOSITE MATERIALS

Annual Technical Report

by

H. M. Rosenberg

January 1977

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Abstract

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The HERA material has a thermal conductivity typical of a heavily-loaded composite and an electrical conductivity which is typical of a metallic alloy. The carbon fibre thermal conductivity both perpendicular and parallel to the fibres is exceedingly low at helium temperatures but it increases rapidly as room temperature is approached. Its electrical resistance increases slightly down to 20 K but it decreases as the temperature is further reduced.

The thermal expansion of carbon fibre composite has been measured down to 20 K. On cooling it expands in the direction of the fibre axis but it contracts perpendicular to the axis and this suggests a method of making a material of high dimensional stability.

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Contents

Introduction

This report consists of results from work which is still in progress. The data are therefore provisional and we are not yet in a position to present a full discussion of the experimental results or a detailed analysis in the light of current theories.

The investigations are as follows:

(1) The thermal and electrical conductivity of HERA polymer-moulded samarium-cobalt magnets from 2 K to room temperature.

(2) The thermal and electrical conductivity of carbon fibre composite materials from 2 K to room temperature.

(3) The thermal expansion of carbon fibre composite materials from 20 K to room temperature.

Chapter 1

A Preliminary Study of the Thermal and Electrical Conductivity of Polymer Moulded Samarium-Cobalt Magnets

by G. R. Bandurek and H. M. Rosenberg

1.1 The Samples and Experiments

Samarium-cobalt powder incorporated in a polymer matrix is a new magnetic composite that has a high coercivity and remanence coupled with a very strong resistance to demagnetization. The material can be moulded to complicated shapes and it can also be machined.

Some samples of this material under the trade name HERA were kindly supplied by Magnetic Polymers Limited, Swindon, Wilts., England, in order that a preliminary study of the thermal and electrical conductivities could be made.

The specimens were in the form of rods of rectangular cross-section 7.63 x 8.95 mm and length 51 mm. The thermal conductivity has been measured in the range 2 K up to 78 K in a standard helium cryostat. Measurements from 78 K to room temperature were made on a thin slice (2 mm) of material between copper plates using a Lees disc experimental arrangement. For the low temperature thermal conductivity measurements clamps were attached to the specimen 10.8 mm apart to which were attached Au/Fe:chromel thermocouples. These were used to measure the temperature gradient along the specimen when heat was generated electrically in a winding attached to one end of the rod. In the Lees disc arrangement the thermocouples were set into the copper plates on either side of the specimen.

The electrical resistance was measured using a standard four-terminal circuit. Clamps were used for the current and potential connections to the specimen.

1.2 Thermal Conductivity

The graph of the thermal conductivity against temperature is shown on a log-log scale in figure 1. From 2 to 10 K the conductivity has a temperature dependence of approximately $T^{1.6}$ but the dependence on T becomes less rapid as the temperature is increased.

The conductivity is rather low. At room temperature it is about $1.5\ Wm^{-1}\ K^{-1}$ and at 10 K it is of the order of $10^{-1}\ Wm^{-1}\ K^{-1}$. The form of the curves and the magnitude of the conductivity is very similar to results which we have obtained for composites of fine metallic powders in an epoxy-resin matrix (e.g. see Araujo and Rosenberg, 1976). For example, in this work the sample of an epoxy-resin/copper composite with a volume concentration of 54.6% of 11 micron diameter spherical particles had a thermal conductivity of about $2 \times 10^{-1}\ Wm^{-1}\ K^{-1}$ at 10 K. An optical microscope observation of the HERA material shows that it is made of irregularly shaped particles most of which are in the range 10-30 microns, with a volume

concentration of around 50%.

It might sound surprising that the conductivity of a composite containing an alloy should be so similar to that of one containing high purity copper (which one might think should have a much higher thermal conductivity). Our previous work, however, has shown that at low temperatures in particular the thermal conductivity of a composite is not very dependent on the intrinsic conductivity of the metallic particles. The dominant effect is due to an acoustic mismatch between the resin and the particles which scatters the lattice vibrations and gives rise to an extra thermal resistance. This acoustic mismatch depends only on the elastic constants of the metal and of its density - and these will not be so very different for the samarium-cobalt and for copper. Hence the thermal conductivities of the two types of material will be similar to one another.

1.3 Electrical Resistivity

The electrical resistivity of the HERA material has been measured from room temperature to 2 K. The results are presented in figure 2. This shows that the general behaviour of the electrical resistivity of the material is very similar to that of a metal. As the temperature is decreased the resistivity decreases quite rapidly but it flattens off to a fairly constant value at temperatures below about 20 K. Contrary to our work on the electrical properties of copper-epoxy composites (which formed the basis of our annual report last year) the HERA materials exhibit quite a reasonable conductivity (of the order of 2×10^4 ohm^{-1} m^{-1}) even at low applied potentials. This would indicate that the polymer used for the matrix in the HERA specimens does not flow so as to inhibit contact between the metallic particles as it did seem to do in our copper specimens. The ratio of the electrical resistance at room temperature to its constant value at low temperature is very small - about 1.3 - which is not surprising since the metallic particles are made of a complicated alloy which has been produced by sintering. This would give rise to a large impurity scattering for the conduction electrons and it would lead to a small resistance ratio. The rather low electrical conductivity over the whole temperature range would suggest that any eddy current problems which might arise when the material is used would not be accentuated in any low temperature applications.

Chapter 2

The Thermal and Electrical Conductivity of Carbon Fibre Reinforced Plastic from 2 K to Room Temperature

by D. J. Radcliffe and H. M. Rosenberg

2.1 The Samples and Experiments

The thermal and electrical conductivities of carbon fibre reinforced plastic have been measured in directions both parallel and perpendicular to the fibre axis in the range from 2 K up to room temperature. The material was the same as that described in the following paper on the thermal expansion of these materials (Pinheiro and Rosenberg) and both HT type (high tensile) and A type (high strength) specimens were investigated.

The conductivities were measured in a conventional helium cryostat. Specimens approximately 3 x 3 mm in cross section and 3 cm long were used and the thermocouples (Au/Fe:chromel) were anchored to the specimen at copper pins about 10 mm apart which were set with epoxy into small holes drilled into the specimen. For measurements from 78 K to room temperature thin slices of material were used in a Lee's disc arrangement in order to minimise the errors due to thermal radiation.

The electrical conductivity was measured using a standard four-terminal circuit. The potential contacts were the same as those used for the heat conductivity measurements.

2.2 Thermal Conductivity

The values of the thermal conductivity of both types of carbon fibre specimens and also the epoxy Epikote 828 are shown on a log-log plot in figure 3. As is to be expected, the conductivity parallel to the fibres is greater than that transverse to the fibres. It will also be noted that there is some difference between the HT and the A type materials. At room temperature the HT type has the higher conductivity and this leads in the longitudinal direction to a value which is approximately twice as great as that for the A type. In the transverse direction this difference is not so marked. Below about 20 K the relative values of the conductivities are reversed so that specimens containing the A type material have the higher conductivity. In the range 5-15 K in all four curves there is evidence for a slight levelling off of the curves which is a reflection of the plateau that occurs in the thermal conductivity of the epoxy resin itself (and indeed of all glassy materials) in this temperature range. Above 20 K the heat conductivity of the composites is higher than the epoxy, becoming about 100 times greater at room temperature. In the liquid helium region the thermal conductivity of the epoxy is between 2 and 8 times greater than that of the carbon fibre material. In fact at about 2 K and below the heat conductivity of the carbon fibre specimens transverse to the fibre axis is as low as (if not lower than) any other thermal conductivity which we have measured (e.g. 30% 11 micron corundum powder in epoxy and 30% 0.5 to 1 micron diameter diamond

powder in epoxy, see Garrett and Rosenberg, 1974).

If the conductivity of the fibres along their axis is deduced from that of the composite by subtracting the conductivity of the epoxy matrix weighted according to its volume fraction, a rough estimate of the phonon mean free path, λ, can be made. Using Slack's (1962) value for the longitudinal velocity of sound and Desorbo and Nicholls' (1958) value of the specific heat of pyrolytic graphite at 3 K, when the dominant phonon scattering mechanism will be crystallite boundaries, one obtains λ = 12 nm for HT and 26 nm for A fibres. Electron micrographs of carbon fibres give crystallite dimensions of 10-30 nm which is in agreement with our estimate.

2.3 Electrical Conductivity

The electrical resistivity of C.F.R.P. was found to be very low (between 8-10 ohm m over the whole temperature range studied) so that it was necessary to use an extremely thin sample to obtain accurate results. Since the form factor could not be measured accurately, measurements of the resistivity of a sample which had dimensions similar to the thermal conductivity samples were taken at room temperature (when a large current could be passed through the sample without heating it up significantly). From these measurements the form factor for the thin sample could be calculated. The resistivity measurements were carried out with helium gas in the sample chamber to minimise the temperature difference set up along the sample.

The results for an A-type sample are shown in figure 4. The resistance increases slightly on cooling below room temperature and it reaches a maximum at about 20 K. Below that temperature it starts to decrease again. The increase in resistance on cooling to 20 K is behaviour which is typical of a semiconductor, but the decrease at lower temperatures must be investigated in more detail before any detailed discussion can be given.

Chapter 3

The Thermal Expansion of Carbon Fibre Reinforced Plastic from 20 K to Room Temperature

by M. de F. F. Pinheiro and H. M. Rosenberg

3.1 The Specimens and Experiments

The thermal expansion of composites made from the two main types of carbon fibre - the high-tensile and the high-strength material has been measured from 20 K up to room temperature. The purpose of this paper is to present the results of some preliminary measurements on this material which are interesting in themselves and which may also have possible cryogenic applications but a detailed analysis will not be presented here.

The specimens were prepared for use by Courtaulds Ltd. from their Graphil fibre and they contained 60% by volume of continuous fibre in an epoxy resin matrix (Epikote 828). Samples were made from the two main types of carbon fibre - HT type (high tensile material) and A type (high strength material) and measurements were taken on both of these. Specimens were cut with a cross section of 5 x 10 mm and length 12 mm. They were mounted in a capacity cell and the change in length as a function of temperature was measured using a standard three-terminal capacity bridge technique (White 1961).

3.2 Experimental results

The change in length relative to the length at room temperature (~293 K) in the direction parallel to the fibres is shown in figure 5. It will be seen that both types of material show an initial contraction down to about 250 K and then they expand at lower temperatures. The overall net expansion of the HT material is about 1.6×10^{-4} down to 20 K. For the A type the net expansion is considerably less, 0.6×10^{-4} down to 20 K.

The fact that these materials expand on cooling has been observed previously (see Pirgon, Wostenholm and Yates, 1973) and although unusual, it is not surprising, because it is known that graphite contracts in the basal plane when it is heated (Nelson and Riley, 1945) and the carbon fibres are composed of oriented graphite chains with the hexagonal axis normal to the fibre axis.

The most interesting thing about these results is the very high dimensional stability of these materials. It is about an order of magnitude or more better than most constructional materials.

The thermal expansion of mixtures of HT and A type fibres in an epoxy matrix is as might be expected, intermediate between the results obtained for the A type and HT type specimens.

Measurements of the change in length perpendicular to the fibre axis show that in this direction these materials contract on cooling in the usual way (figure 6). The magnitude of the effect is a net

change of -6×10^{-3} on cooling to 20 K and it is not particularly small compared with other materials.

3.3 Improving the dimensional stability

The fact that on cooling these materials show an expansion along the fibre direction and a contraction across the fibres suggest that a material containing a suitable combination of fibres laid at right angles to one another should have practically no dimensional change on cooling to 20 K or lower temperatures. It is possible however that the strains set up in such a material might be large enough to lead to cracking and this must be investigated. Another possibility would be to use a lower concentration of parallel A-type fibres in the resin. This should also give dimensional stability but it might be at the expense of a deterioration of the mechanical properties.

3.4 Acknowledgements

We are most grateful to Courtaulds Limited for providing us with the specimens for these experiments. This work is being done during the tenure of a scholarship by M. de F. F. P. from the Instituto de Alta Cultura, Portugal.

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Captions to Figures

Figure 1 The thermal conductivity of HERA, polymer-moulded samarium-cobalt magnet material from helium temperatures to room temperature.

Figure 2 The electrical resistivity of HERA, polymer-moulded samarium-cobalt magnet material from helium temperatures to room temperature.

Figure 3 The thermal conductivity from helium temperatures to room temperature of carbon fibre reinforced plastic for HT (high tensile) and A (high strength) materials. Curves are plotted for conductivities both parallel and perpendicular to the fibre axis. The thermal conductivity is also shown for an unfilled epoxy-resin sample of the same type as that used in making CFRP.

Figure 4 The electrical resistivity of A-type (high strength) carbon fibre reinforced plastic from helium temperatures up to room temperature.

Figure 5 The relative change in length, compared to the length at room temperature, of HT and A-type carbon fibre reinforced plastic. The measurements are taken parallel to the fibre axis.

Figure 6 The relative change in length, compared to the length at room temperature, of HT and A-type carbon fibre reinforced plastic. The measurements are taken in a direction perpendicular to the fibre axis.

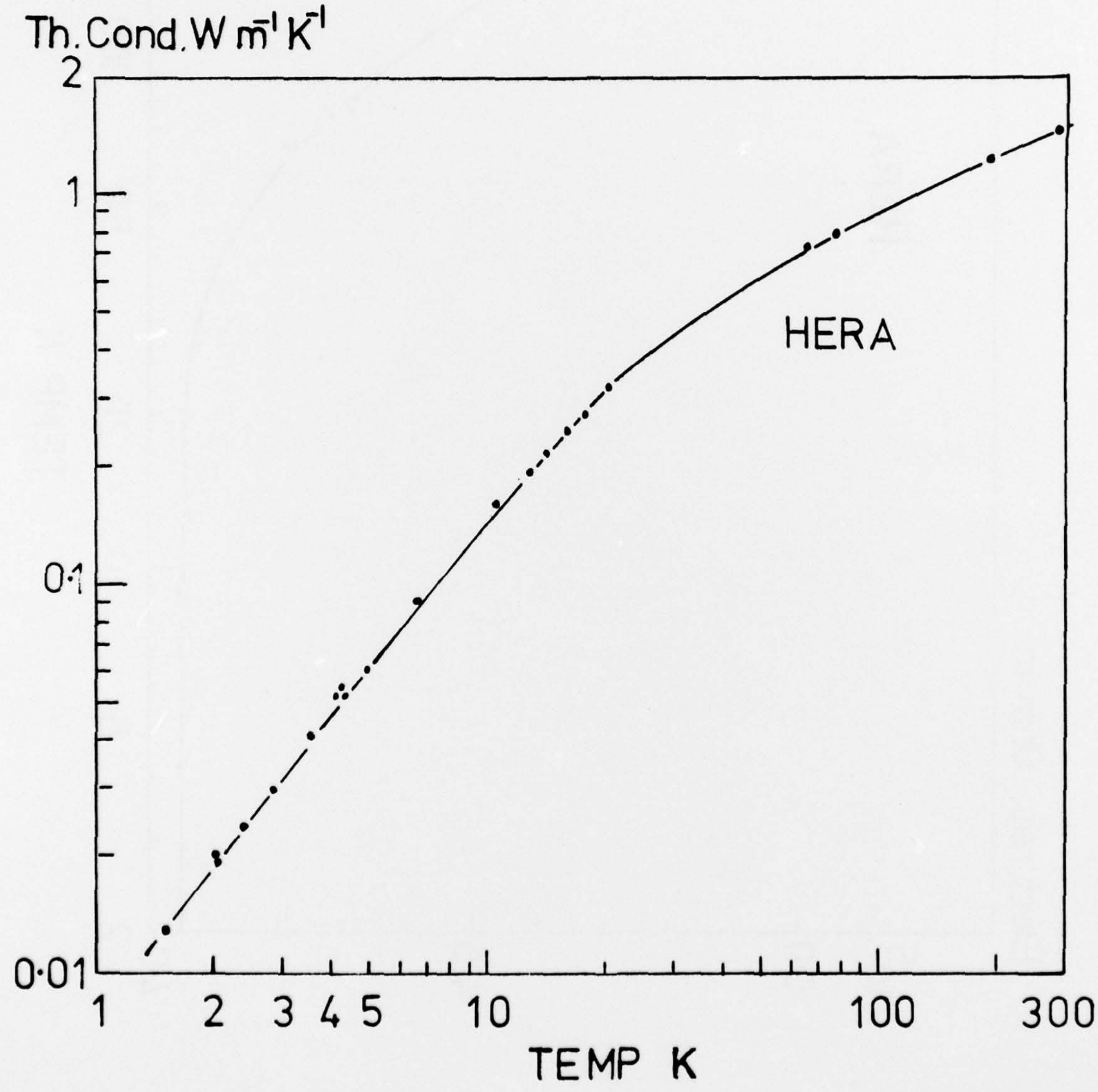


FIG.1

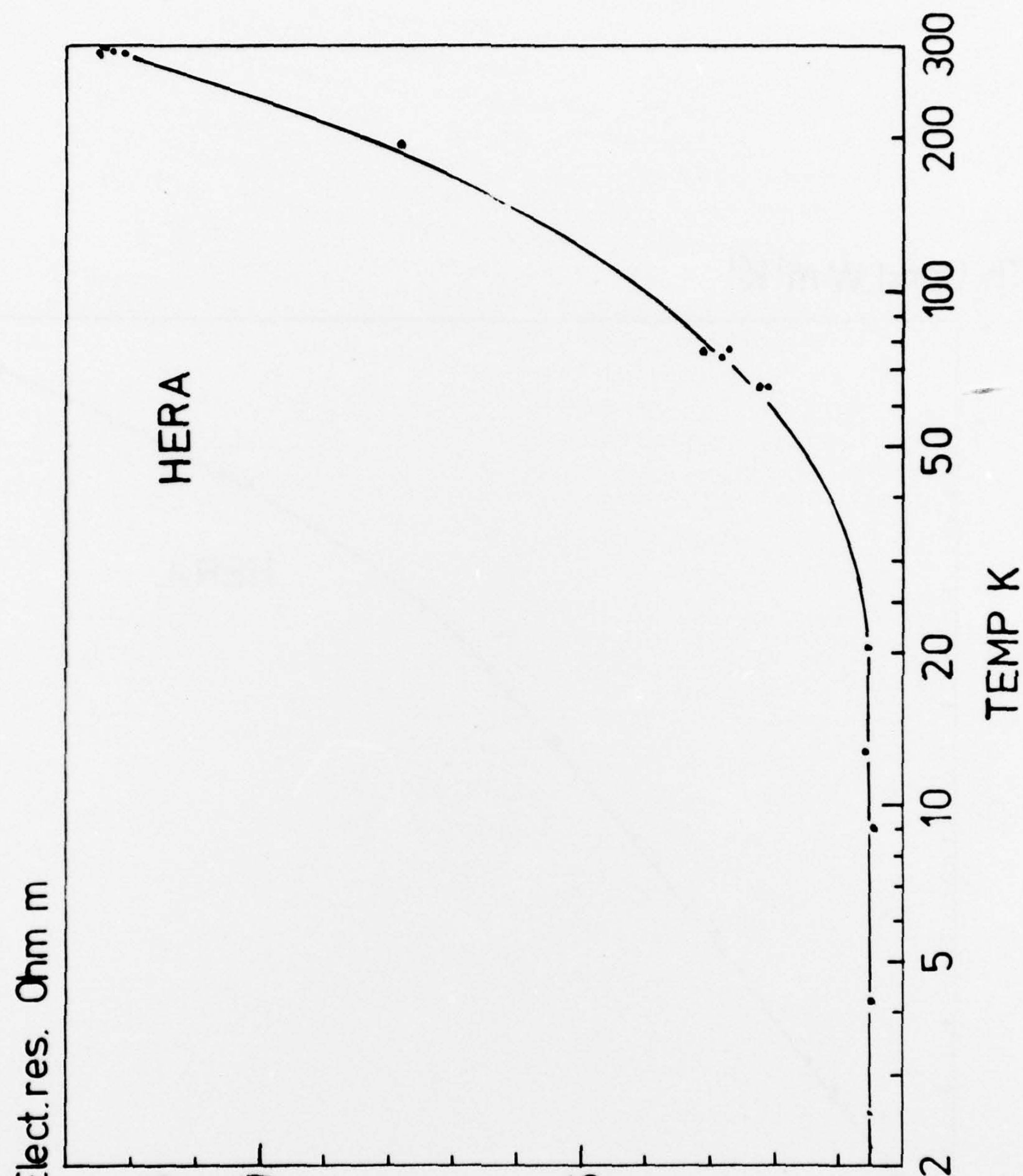
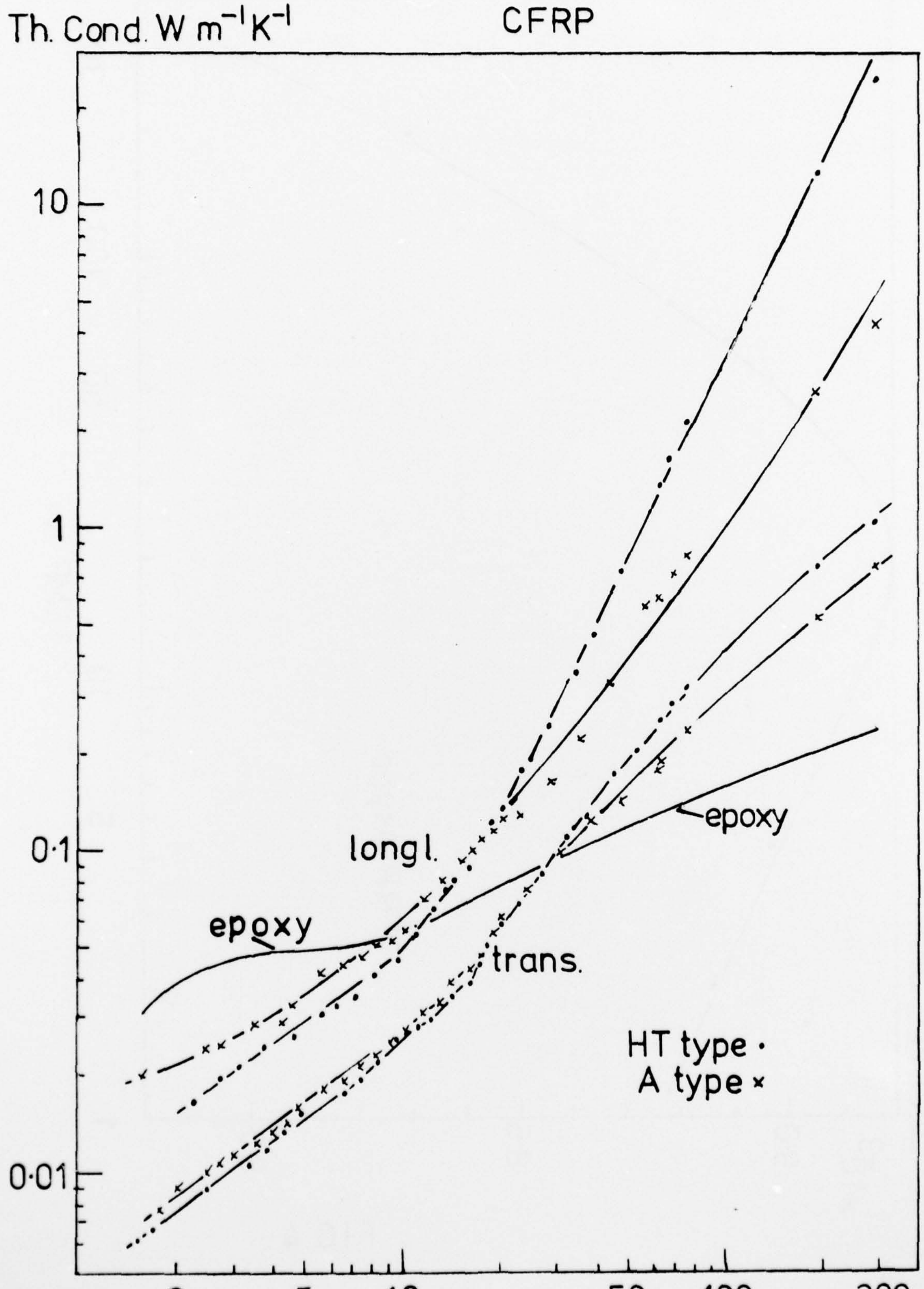

Elect. res. Ohm m
$\times 10^{-5}$
5·0
4·5
4·0
HERA
2
5
10
20
50
100
200
300
TEMP K

FIG. 2



Th. Cond. W m⁻¹K⁻¹
CFRP
10
1
0·1
0·01
longl.
epoxy
epoxy
trans.
HT type •
A type ×
2
5
10
50
100
300
TEMP K

FIG. 3

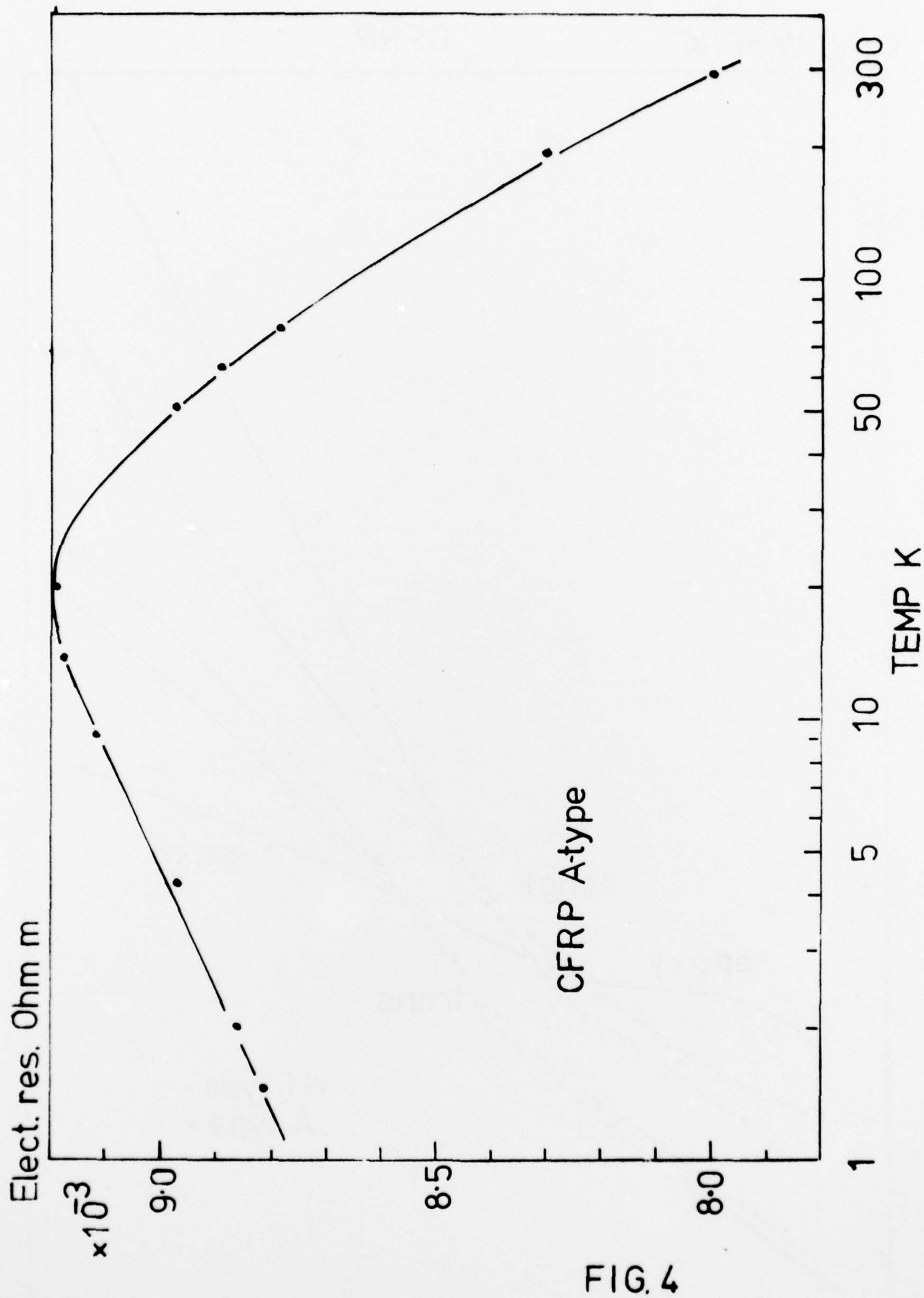


FIG. 4

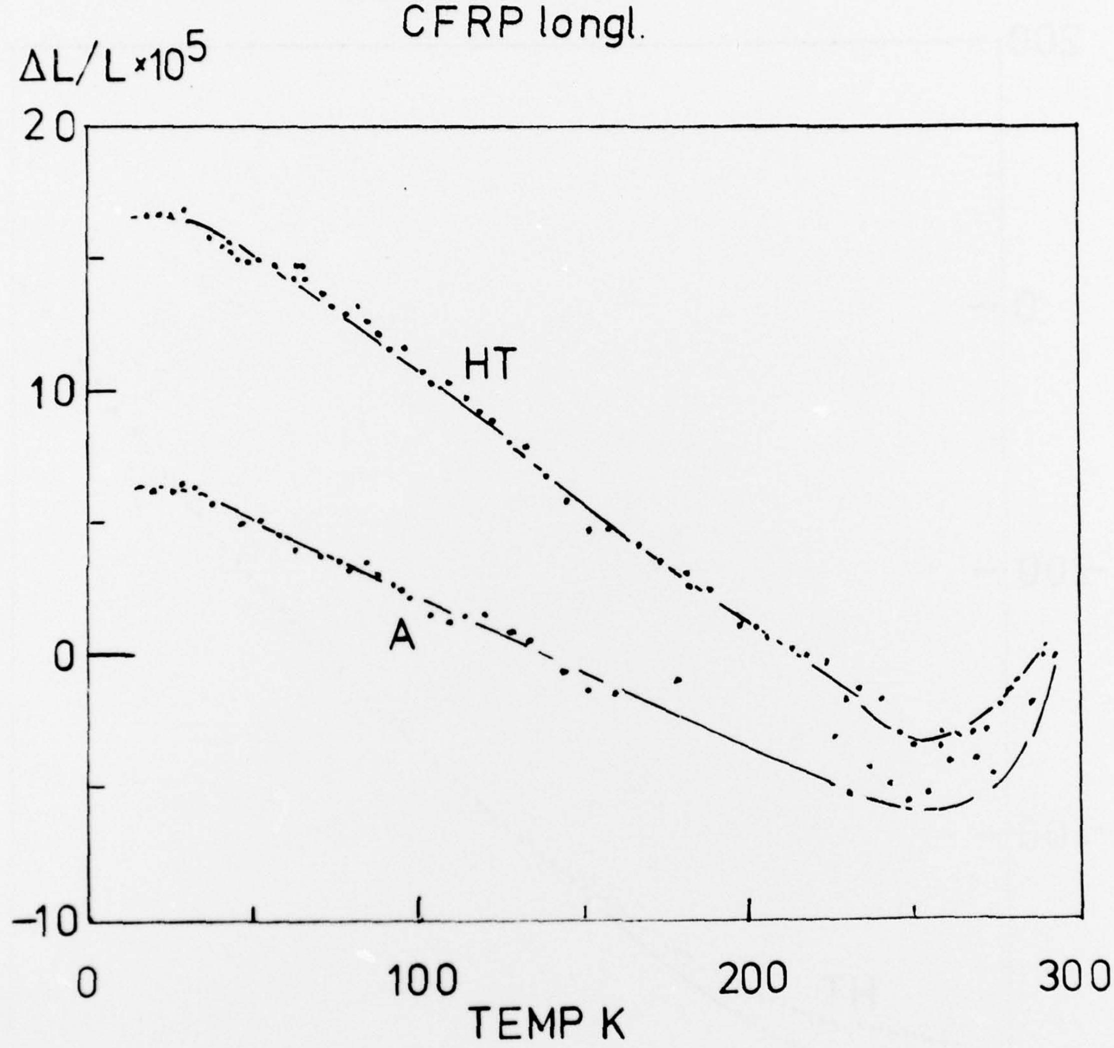


FIG.5

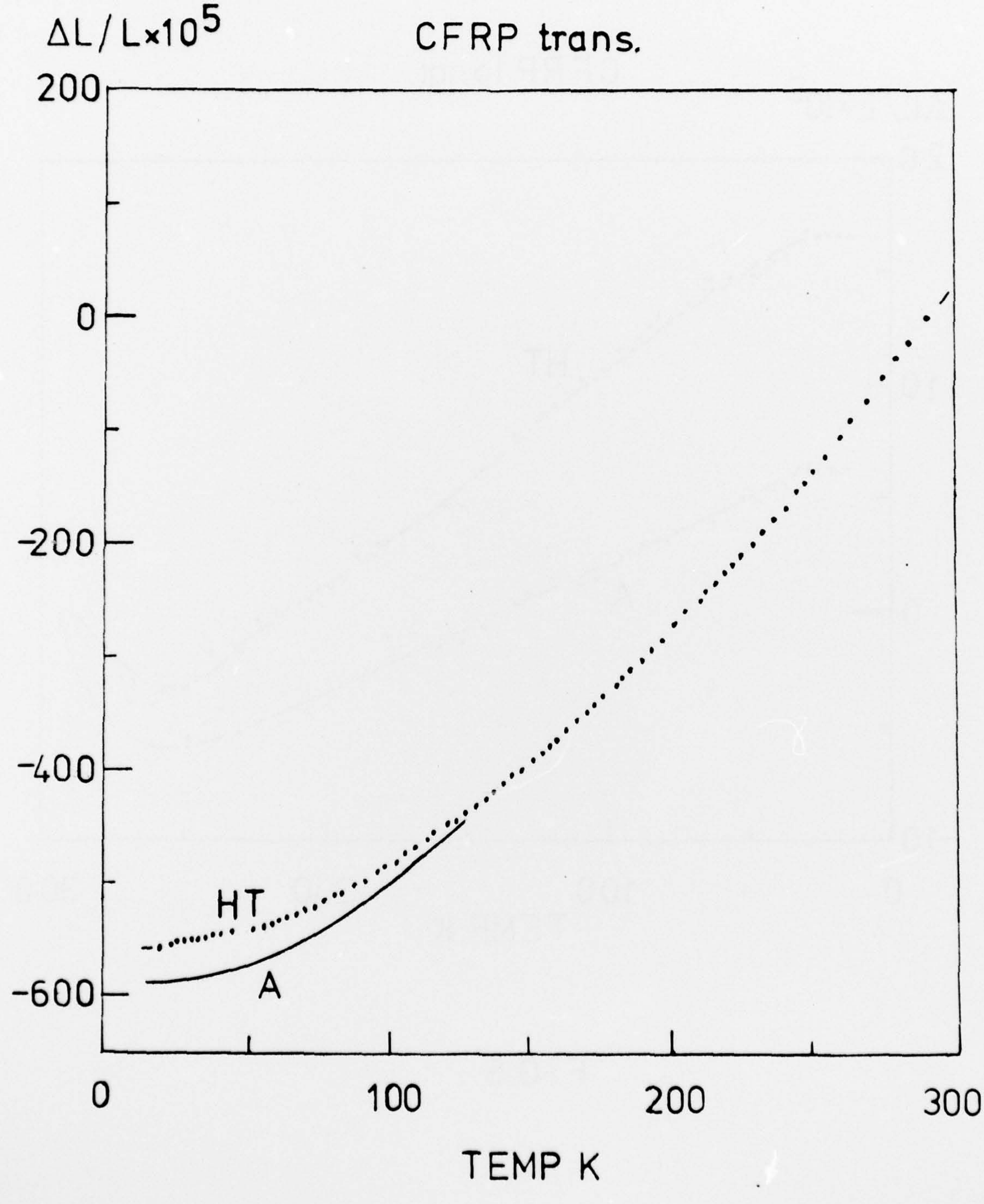


FIG.6